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A RADON (THORON) DAUGHTER PERSONAL ALPHA-DOSIMETER OF THE PASSIVE TYPE USING A DIFFUSED-JUNCTION DETECTOR AND AN ELECTROSTATIC COLLECTOR

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ELLIOT LAKE LABORATORY

MAY 1984

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30 pp

MINERALS RESEARCH PROGRAM
MINING RESEARCH LABORATORIES
DIVISION REPORT MRP/MRL 84-60 (TR)

SEP 24 1984

MICROMEDIA

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J. Bigu* and A. Frattini**

ABSTRACT

A solid-state alpha-dosimeter has been designed and found to be suitable for personal and environmental radon/thoron daughter monitoring. The dosimeter basically consists of an electrostatic collector and an alpha-particle counting system with spectroscopy capabilities. The sensitive volume (~20 cm^3) of the electrostatic collector consists of a cylindrically-shaped metal wire screen and a diffused-junction silicon alpha-detector covered with a thin aluminized mylar sheet. A DC voltage (~450 V) is applied between the wire screen and the mylar sheet, the latter held at negative potential relative to the metal screen. Data can be retrieved during or after sampling by means of a micro-computer (Epson HX20) via a RS-232 communication interface unit. The dosimeter has been calibrated in a large (26 m^3) radon/thoron test facility. A linear relationship was found between radon gas concentration and radon daughter Working Level, and the dosimeter's alpha-count. The dosimeter is mounted on top of an ordinary miner's cap lamp battery and is ideally suited for personal monitoring in underground uranium mines and other working areas. The dosimeter presented here is a considerably improved version of an earlier prototype.

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DOSIMÈTRE ALPHA INDIVIDUEL DE TYPE PASSIF POUR MESURER LES PRODUITS DE FILIATION DU RADON (THORON) AU MOYEN D'UN DÉTECTEUR À JONCTION DIFFUSÉE ET D'UN COLLECTEUR ÉLECTROSTATIQUE

par

J. Bigu* et A. Frattini**

RÉSUMÉ

Les auteurs ont conçu et testé un dosimètre alpha à semiconducteur qui sert à mesurer les concentrations en produits de filiation du radon (thoron) chez les individus et dans l'environnement. Le dosimètre se compose essentiellement d'un collecteur électrostatique et d'un dispositif de comptage des particules alpha avec capacité spectroscopique. La partie sensible (volume 20 cm^3) du collecteur électrostatique est constituée d'un treillis métallique de forme cylindrique et d'un détecteur alpha à silicium à jonction diffusée couvert d'une mince feuille de mylar aluminisé. Une tension CC (450 V) est appliquée entre le treillis et la feuille de mylar, laquelle est chargée négativement. Les données peuvent être recueillies pendant ou après l'échantillonage au moyen d'un micro-ordinateur (Epson HX20) via un appareil de transmission de données RS-232. Le dosimètre a été étalonné dans une large enceinte (26 m^3) servant à l'expérimentation sur le radon (thoron). Une relation linéaire a été établie entre la concentration de radon et l'unité alpha (Working Level) des produits de filiation du radon d'une part, et le compte alpha du dosimètre d'autre part. Le dosimètre est monté au sommet de l'accumulateur qui alimente un casque de mineur ordinaire et convient parfaitement à la mesure individuelle dans des mines d'uranium souterraines et dans d'autres milieux de travail. Le dosimètre présenté ici est une version largement améliorée d'un prototype antérieur.

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INTRODUCTION

Personal α-particle dosimeters have been designed to determine the exposure of miners and other occupational workers to α-radiation from uranium mines and related processing and refining industries.

The use of personal α-dosimeters provides the most direct way of estimating personal radiation exposure. The need for a reliable α-dosimeter has been recognized for some time. There is, however, no single personal dosimeter that combines the broad spectrum of practical requirements including: being inexpensive to build; rugged; simple to maintain and retrieve information from; long-time reliability; and, in addition, responsive to the growing need for discriminating the thoron progeny present in variable amounts in some Canadian uranium mines.

The choice of one dosimeter type over another must be based on a number of factors including long-term reliability, ruggedness, initial investment and routine maintenance, ease of operation, and data processing and retrieval. In spite of the obvious benefit of personal α-dosimeters in the estimation of exposure of uranium miners their use is not widespread. There are only a few mines where α-dosimeters are employed on a routine basis.

Personal α-dosimeters are time-integrating devices that usually consist of a highly efficient pump, a sampling head housing an adequate filter, and an α-detector facing the 'active' side of the filter. Air is continuously sampled through the filter where the radioactive products are deposited. Alpha-particles emitted by the radioactive products are detected by an appropriate detector, e.g., a thermoluminescent detector, such as LiF or CaF_2:Dy, a track-etch detector such as cellulose nitrate, or a silicon diffused-junction or surface-barrier detector.

Extensive research and development and laboratory and field testing

have been conducted on the above dosimeter prototypes by several authors (1-9). These dosimeters are of the so-called active type, i.e., they use a pump to sample airborne particulates and a filter where the radioactive products are deposited for measurement. Broadly speaking, there are two major difficulties associated with the use of active dosimeters. These difficulties which are mainly related to the sampling pump are:

a) Lack of pump reliability associated with pump mechanical design, and wear and tear;

b) Need for portable power supply to power the pump during the sampling period.

Item (a) leads to appreciable uncertainty in the total volume of air sampled and the constancy of the volumetric flowrate of the dosimeter during the sampling period. This in turn substantially limits the accuracy with which radiation levels can be determined. Item (b) reduces the useable sampling period and increases the weight and size of the dosimeter.

Because of some of the drawbacks of active dosimeters, a different kind of dosimeter which eliminates the need for a sampling pump and collecting filter has been investigated. This type of dosimeter is known as the passive type. Comparatively speaking, little work has been done in the field of passive α-dosimeters. In principle, a passive dosimeter can make use of the same detection system as that of an active dosimeter. However, there are some drawbacks associated with dosimeters of the passive type, as discussed below.

This paper presents experimental data regarding the design, characteristics and performance of a passive α-dosimeter developed under contract (CANMET, Energy, Mines and Resources Canada) with Alpha-NUCLEAR (Toronto). The dosimeter uses a diffused-junction silicon detector and an electrostatic collector as detection system and is a considerably improved version of an earlier prototype (10). The instrument is battery operated. Calibration of the dosimeter was carried out in a radon/thoron test facility, RTTF, for short (11).

DESCRIPTION OF THE PASSIVE ALPHA-DOSIMETER

The solid-state passive dosimeter prototype basically consists of an electrostatic collector and an alpha-particle counting system. The electrostatic collector consists of a cylindrically shaped metal wire screen and a diffused-junction (DJ) silicon α-detector (400 mm² active surface area, 22.9 mm diameter) covered with a thin aluminized mylar sheet. The screen has been designed so that a membrane sleeve can easily be mounted over the screen on a bottom flange. The screen, membrane and flange are sealed around the edges to eliminate air leaks, and form a well defined sensitive volume or chamber for radioactivity sampling purposes. The membrane filters out the decay products of radon, thoron and other unwanted airborne particulates including water vapour.

A DC voltage is applied between the wire screen and the aluminized mylar; the latter is held at negative potential (∿-500 V) relative to the screen.

The operating principle of the electrostatic collector is as follows. Radon and/or thoron from a radon(thoron)/radon(thoron) daughter mixture passes through the membrane whereas the decay products are removed by the membrane. The freshly filtered radioactive gas decays into its decay products in the sensitive volume of the electrostatic collector. Alpha particles, positively charged, are accelerated towards the negatively charged mylar sheet and into the DJ detector where they are counted. Furthermore, and most importantly, decay products of radon and/or thoron initially formed in a positively charged state, e.g., RaA, in the sensitive volume of the collector are attracted to, and deposited on, the mylar sheet from where they decay and are detected by the DJ detector.

The application of a DC voltage between the wire screen and the mylar

sheet combined with the wire screen/membrane mechanical configuration serves a dual purpose, namely: it considerably enhances the collection efficiency of the electrostatic collector while at the same time it clearly defines a sensitive volume for sampling purposes, to which the collected decay products can be referred to.

The signal from the DJ detector is fed to a charge-sensitive pre-amplifier followed by a voltage amplifier with a gain of 10. Pulse height analysis can be realized by a three threshold comparator/counter circuits. A CMOS microprocessor continuously scans three threshold counters, updating internal 24 bit registers each time a counter is incremented. The threshold level of the three counters is as follows. Level C accepts all pulse energies above the base line noise, i.e., α-particles from the decay of RaA (6.00 MeV), ThC (6.07 MeV), RaC' (7.69 MeV), and ThC' (8.78 MeV). Level B accepts pulses from RaC' and ThC', i.e., energy from slightly below 6 MeV to about 10 MeV. Level A accepts high energy pulses only intended for ThC' counting. For simplicity, the threshold levels will be referred to as channels C, B and A, respectively. The α-count collected is stored in a memory unit. Data can be retrieved at any desired time during or after the sampling period by means of a microcomputer (Epson HX20) via a RS-232 communication interface unit. The dosimeter has α-spectroscopy capabilities. Thus, the signal from the instrument can be fed directly into a multi-channel analyzer for α-spectrum analysis.

The α-count measured by the DJ detector and associated electronic circuitry is directly related to the radioactive gas concentration in the sensitive volume, and hence to the atmospheric concentration. The α-count can also be related to the daughter decay product level, i.e., Working Level (WL), by proper calibration of the dosimeter or if certain assumptions are made. Data analysis is relatively straightforward for radon/radon daughter atmospheres

only. Analysis is considerably more difficult in the presence of radon/thoron mixtures. The theoretical treatment of these cases is dealt with elsewhere (12). The dosimeter microprocessor controls, in conjunction with the computer, all the electronic and mechanical operations of the instrument including the cap lamp (see below).

The dosimeter has been designed primarily for field use, e.g., underground uranium mines and other uranium-related industries and work locations. It is small in size (14 cm x 4.5 cm x 6 cm) and is mounted, and powered, on a conventional miner's cap lamp MSHA battery (approximately 13 cm x 4 cm x 16 cm). The sensitive volume of the dosimeter has been ruggedized carefully by a specially designed aluminum case, provided partly with circular holes ($\sim$ 2 mm diameter) to allow free passage of radon gas through the dosimeter's membrane. The electronic circuitry is equally protected by the same case, but separated from the sensitive volume of the dosimeter by an adequate steel plate. Figure 1 shows a view of the dosimeter.

EXPERIMENTAL PROCEDURE

The response of the dosimeter to known radon gas concentrations and radon daughter concentrations was investigated in the RTTF. Radon gas concentration in the RTTF was monitored using standard grab-sampling techniques, radon gas monitors of the passive type Model alpha-Meter 400, in conjunction with an external automated data logger, manufactured by Alpha-NUCLEAR (Toronto), and a continuous monitoring system, using a scintillation cell, designed and calibrated at our laboratories. Radon daughter Working Levels, WL(Rn), were determined using several calibrated radon daughter continuous monitoring systems including several Model RGA-400 units and one Model WLM-300 unit manufactured by EDA Instruments (Toronto), and a continuous sampling β-detector

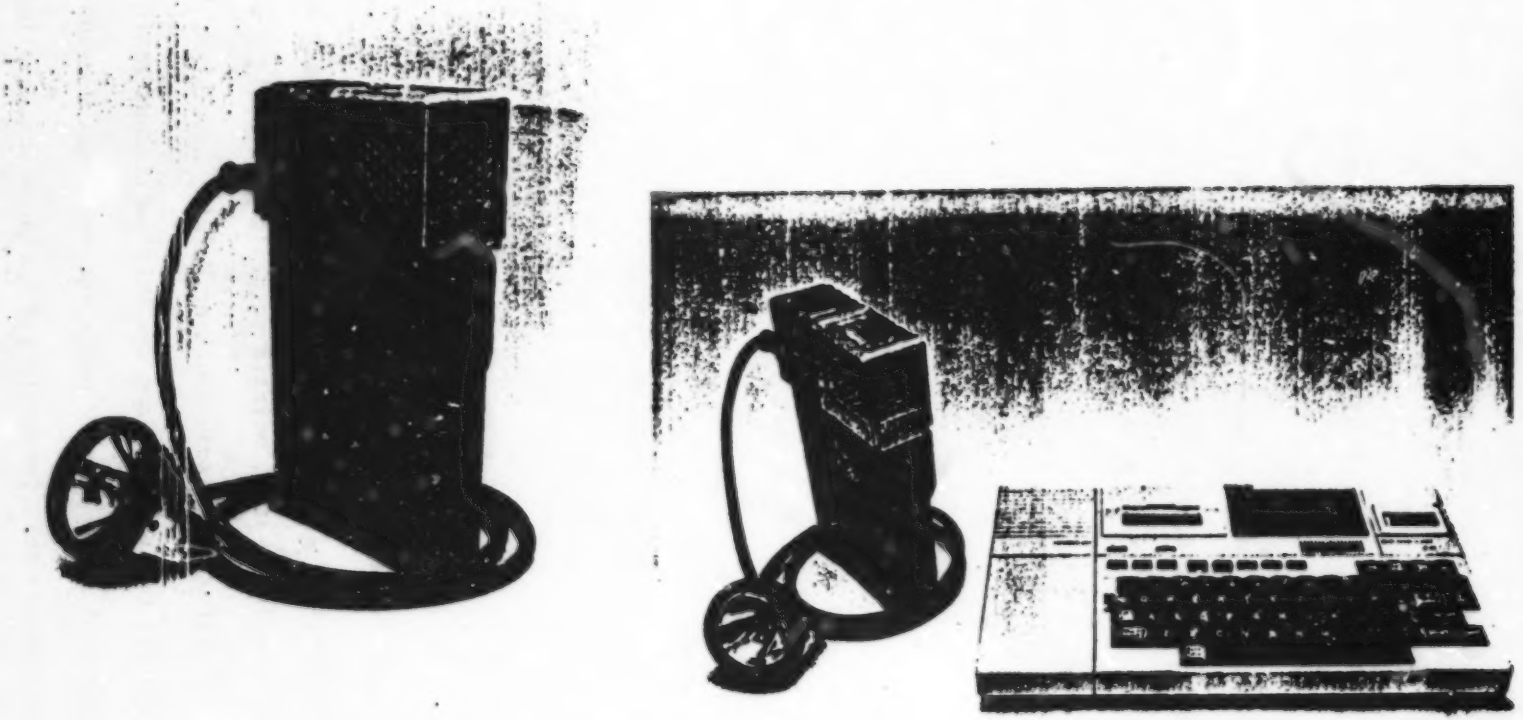

Fig. 1 - Passive dosimeter mounted on miner's cap lamp battery, and computer system. Interface cable not shown.

of the G.M. pancake type designed and calibrated at our laboratories.

Tests were conducted preferentially at low aerosol concentrations ($\sim 1.0 \times 10^3$ cm^{-3}) and within the temperature range 19-25°C. A series of tests were carried out under constant relative humidity conditions (30-40%) at different radon gas concentrations, [Rn] for short, and WL(Rn). In a different series of tests [Rn] and WL(Rn) were maintained constant within ± 10% and the relative humidity was varied from about 30% to ~100%. All experiments were conducted using a membrane manufactured by Chase-Walton Elastomer Inc. (U.S.A.) which has a permeability to radon gas and water vapour of 1.2×10^{-5} cm^2/s and 4.6×10^{-6} cm^2/s, respectively, as determined by Holub (13). Testing of instrumentation under low relative humidity and low aerosol concentration is very important because radon daughters plate-out becomes severe under these conditions and quite sensitive to small changes in the above variables.

Accumulated α-count in channels C, B and A, and calculated accumulated α-count in C-B, during the sampling period and 3.5 h after the end of sampling, including sampling, were plotted versus [Rn] and WL(Rn). The data were fitted to a straight line by linear regression analysis. It should be noted that for radon/radon daughters atmosphere, the contribution from channel A is negligible, as expected. It should also be noted that the α-count in channel C corresponds to the total radon daughter α-count, i.e., RaA + RaC', whereas the α-count in channel B, and channel C minus channel B (i.e., C-B), corresponds, respectively, to RaC' and RaA.

RESULTS AND DISCUSSION

Data pertaining to the α-count in different dosimeter channels versus [Rn] and WL(Rn) are given in Fig. 2 to 7 and Fig. 8 to 13, respectively. Linear regression coefficients (slope, intercept and correlation coefficient)

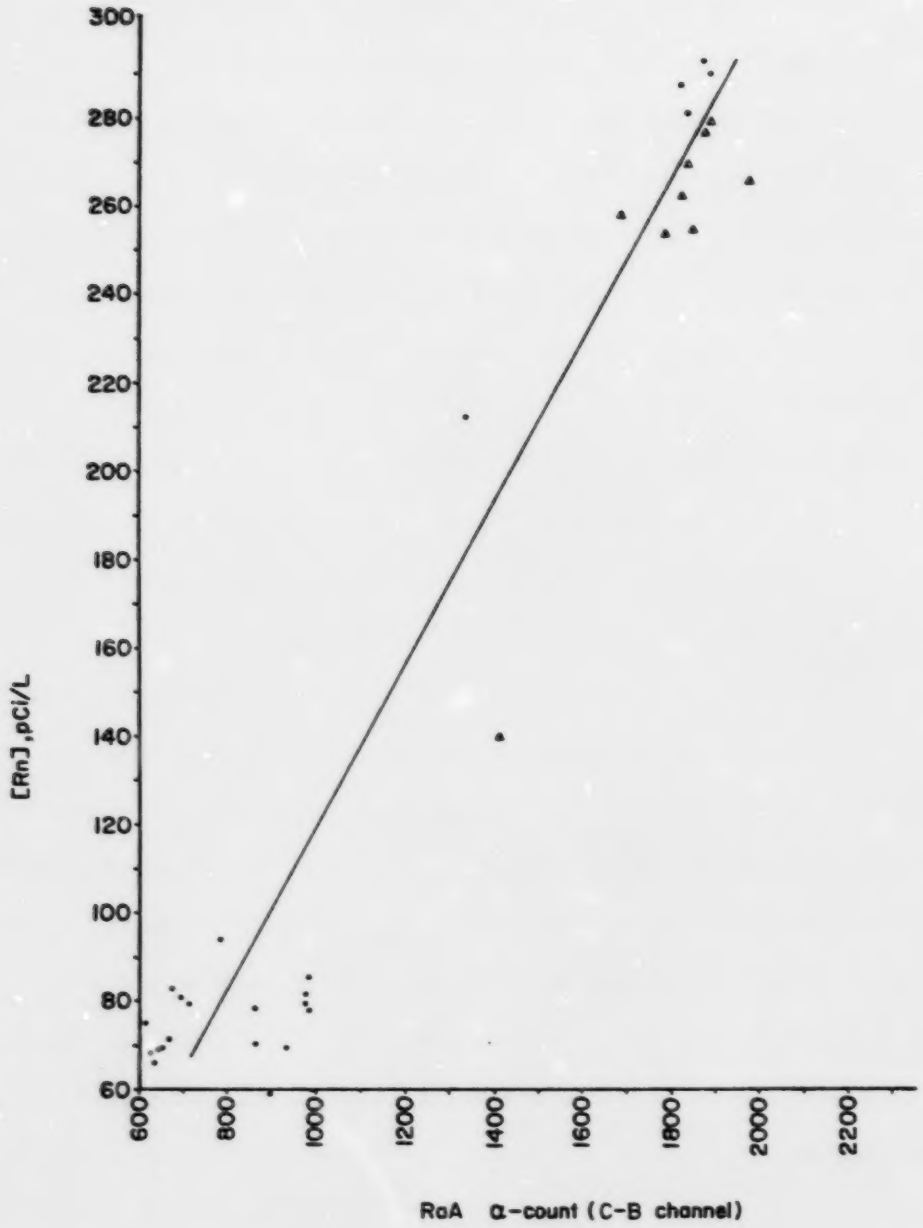

Fig. 2 - Dosimeter RaA α-count versus radon gas concentration. Dosimeter reading taken at the end of sampling.

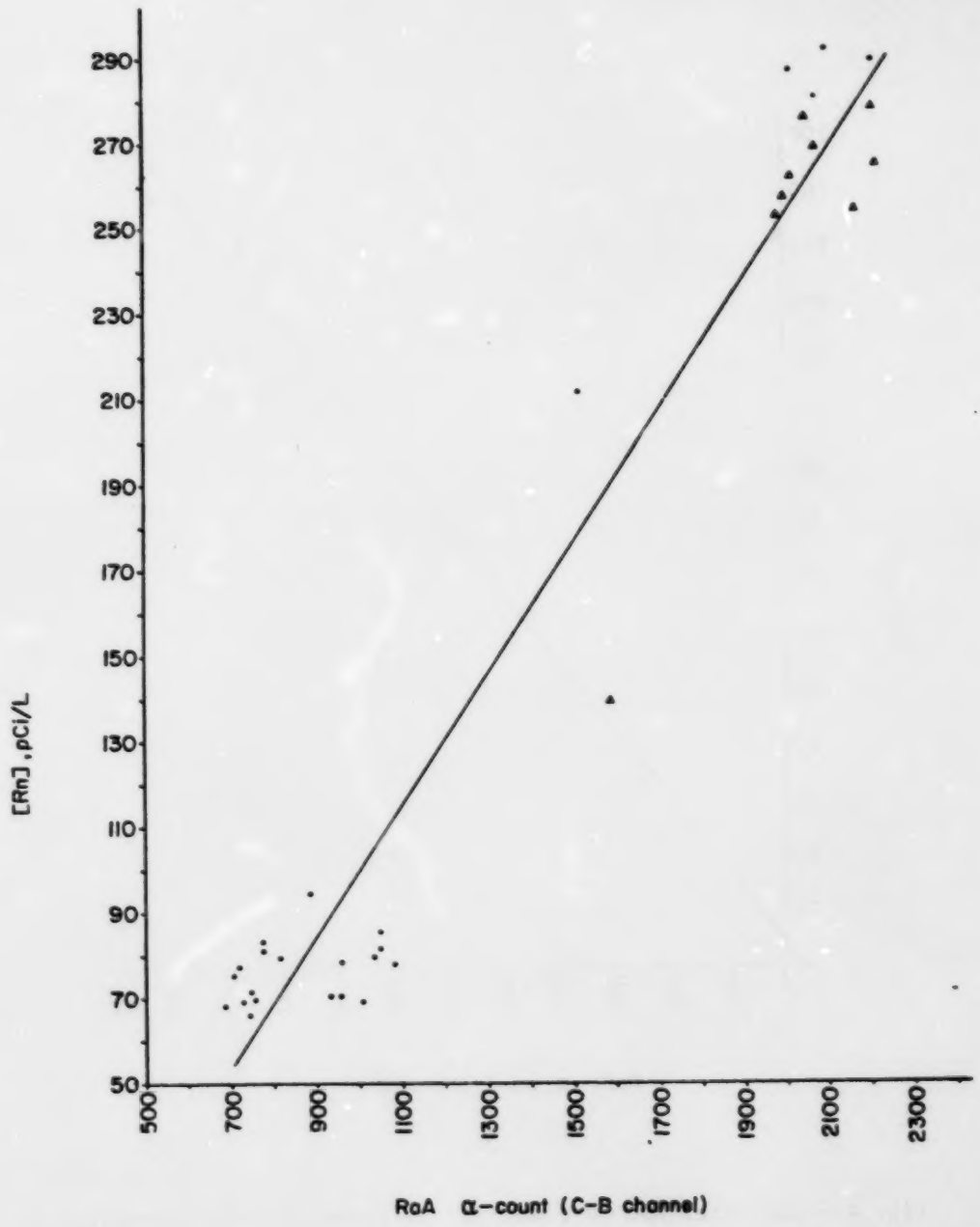

Fig. 3 - Dosimeter RaA α-count versus radon gas concentration. Dosimeter reading taken 3.5 h after sampling.

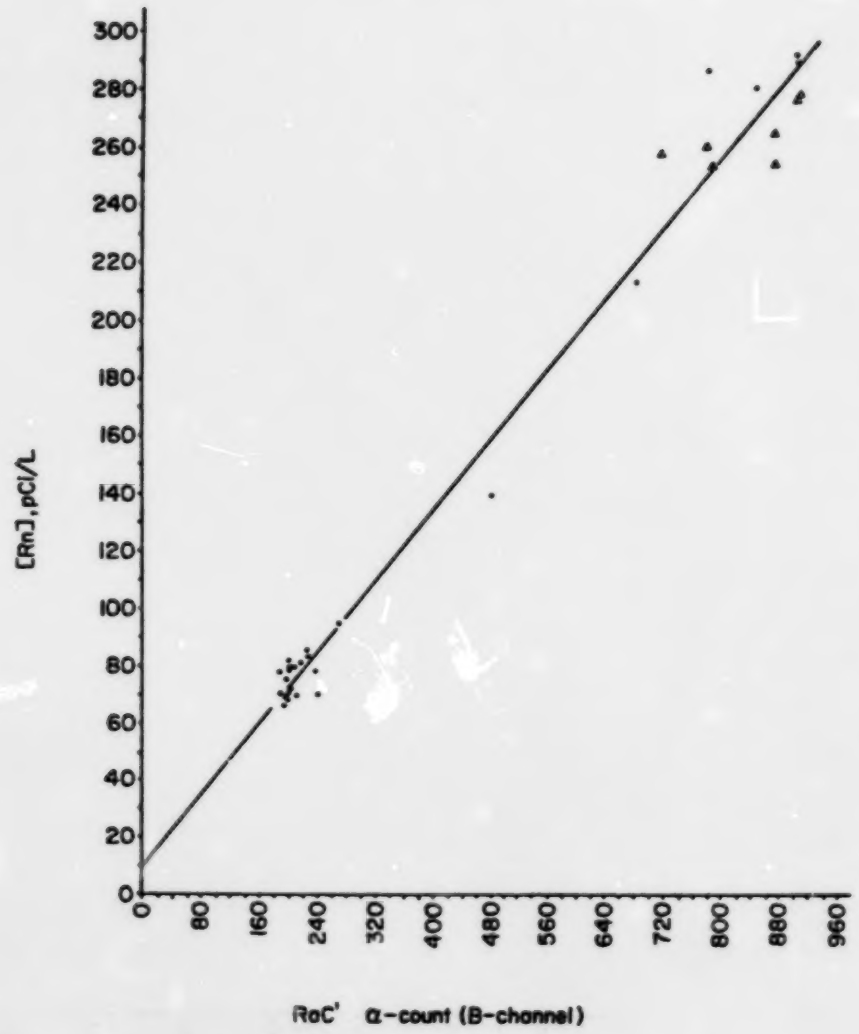

Fig. 4 - Dosimeter RaC' α-count versus radon gas concentration
Dosimeter reading taken at the end of sampling.

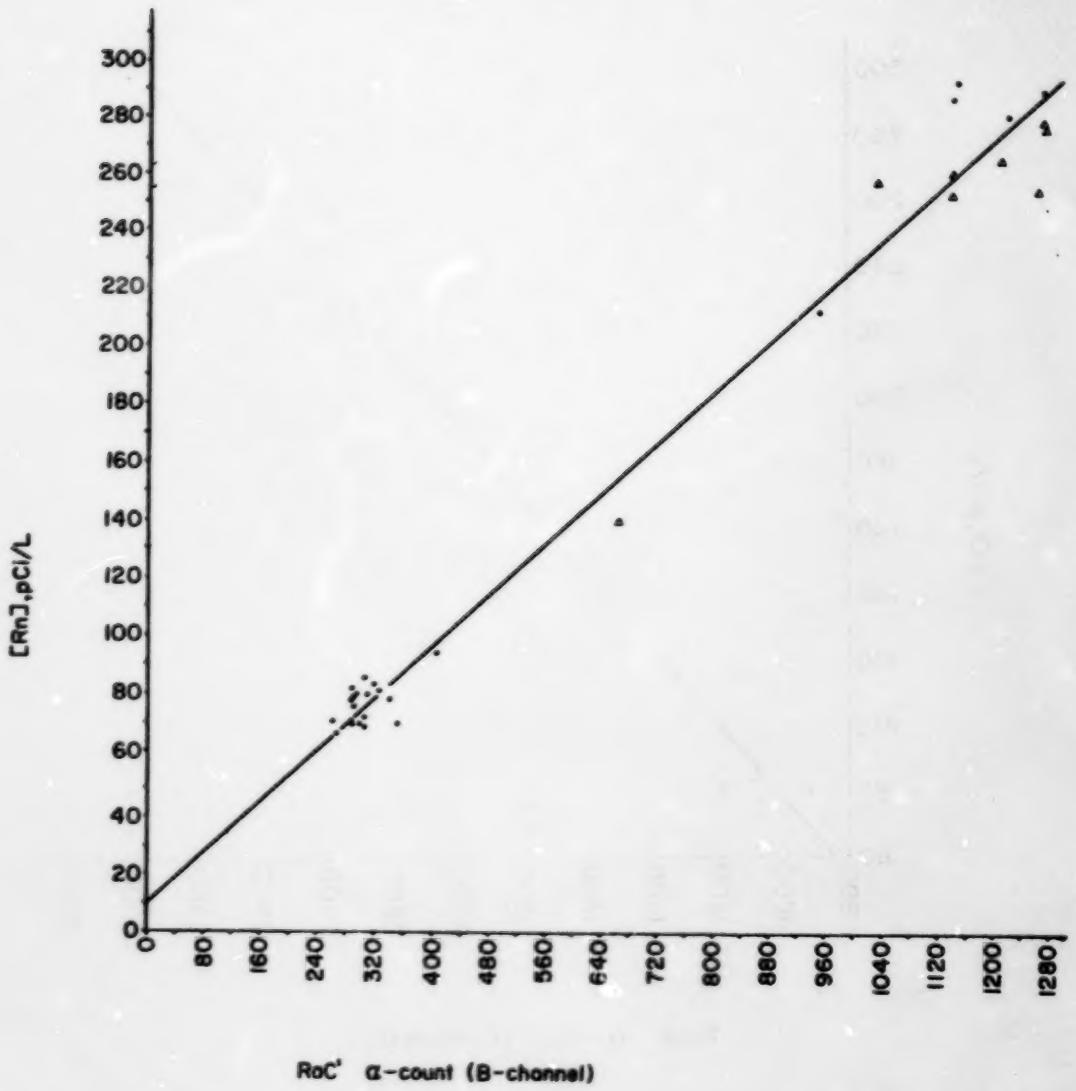

Fig. 5 - Dosimeter RaC' α-count versus radon gas concentration. Dosimeter reading taken 3.5 h after the end of sampling.

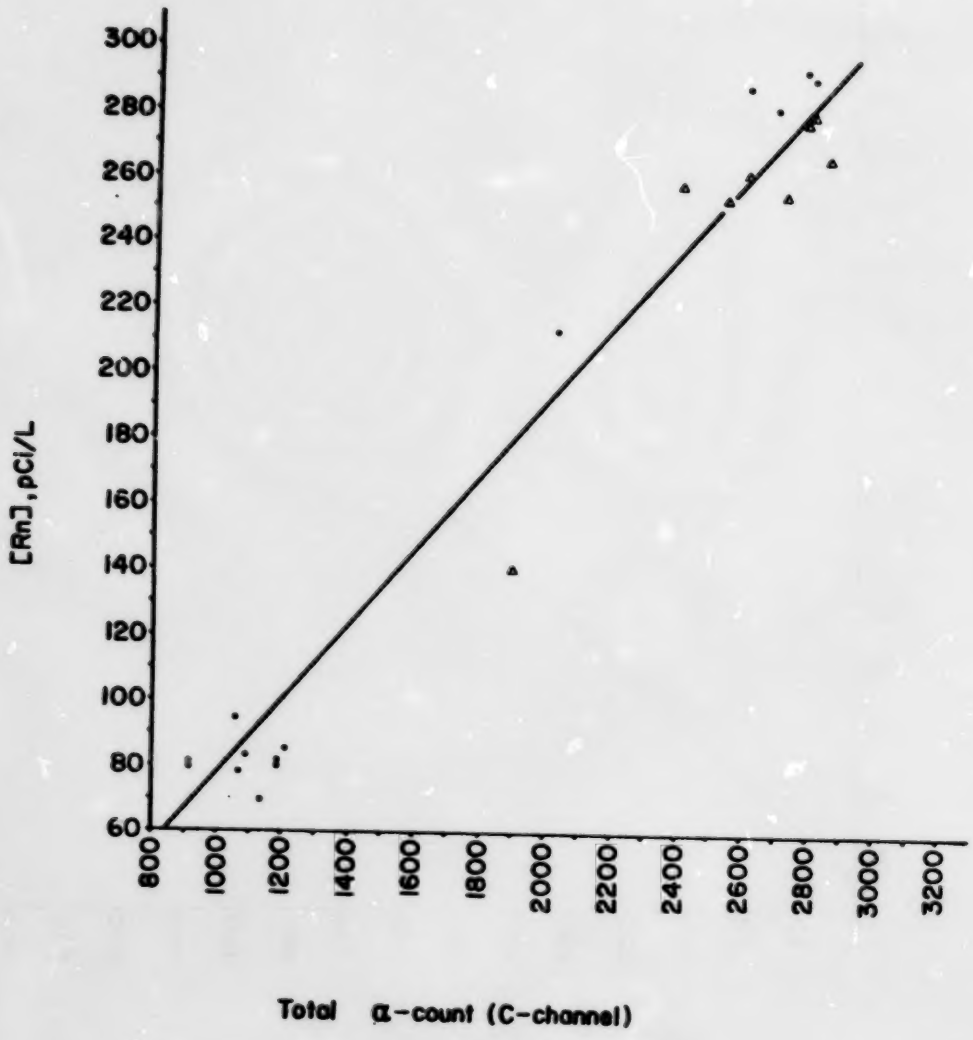

Fig. 6 - Dosimeter total α-count versus radon gas concentration. Dosimeter reading taken at the end of sampling.

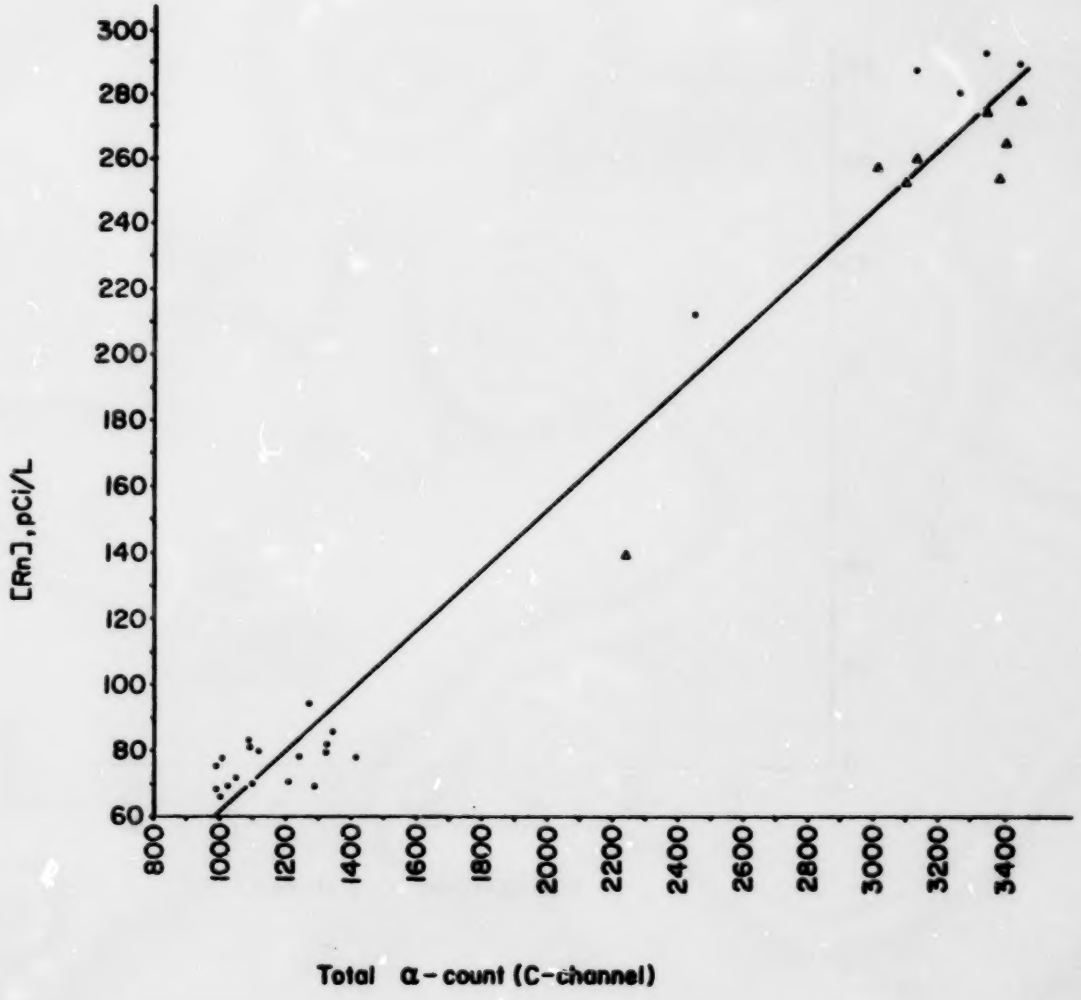

Fig. 7 - Dosimeter total α-count versus radon gas concentration. Dosimeter reading taken 3.5 h after the end of sampling.

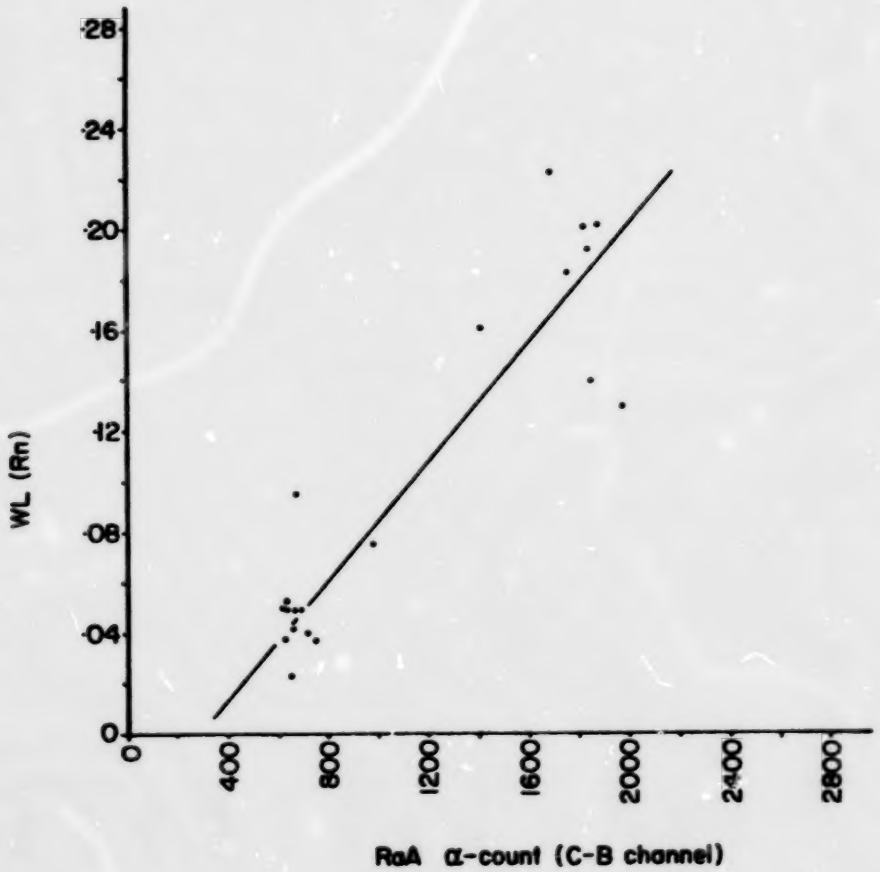

Fig. 8 - Dosimeter RaA α-count versus Working Level.
Dosimeter reading taken at the end of sampling.

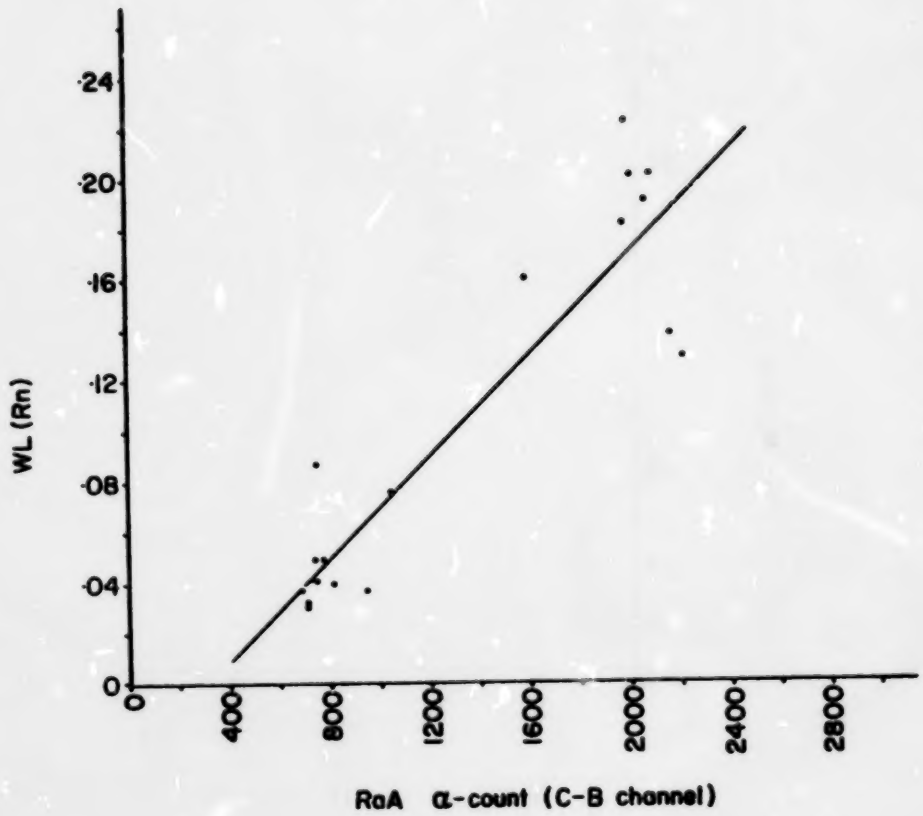

Fig. 9 - Dosimeter RaA α-count versus Working Level.
Dosimeter reading taken 3.5 h after sampling.

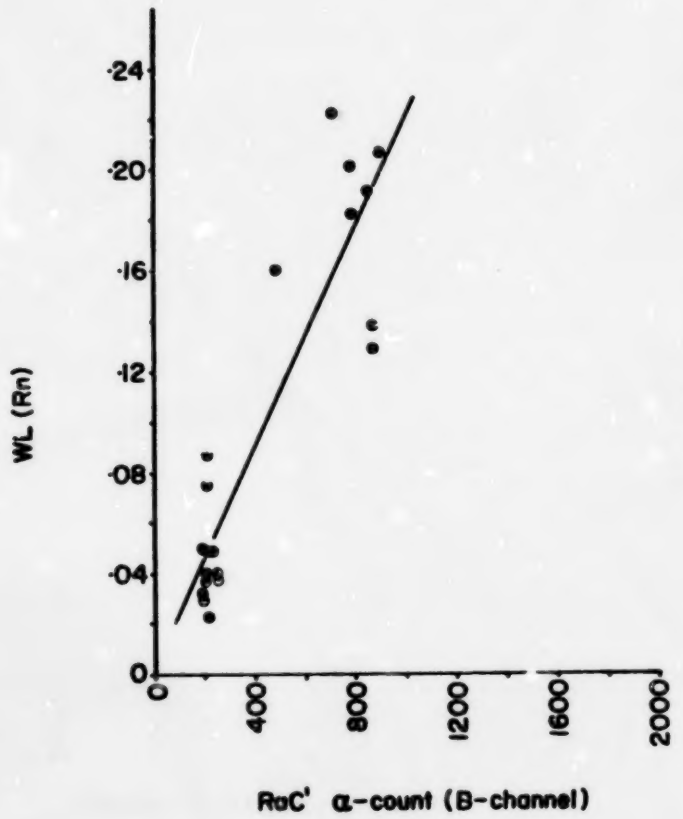

Fig. 10 - Dosimeter RaC' α-count versus Working Level.
Dosimeter reading taken at the end of sampling.

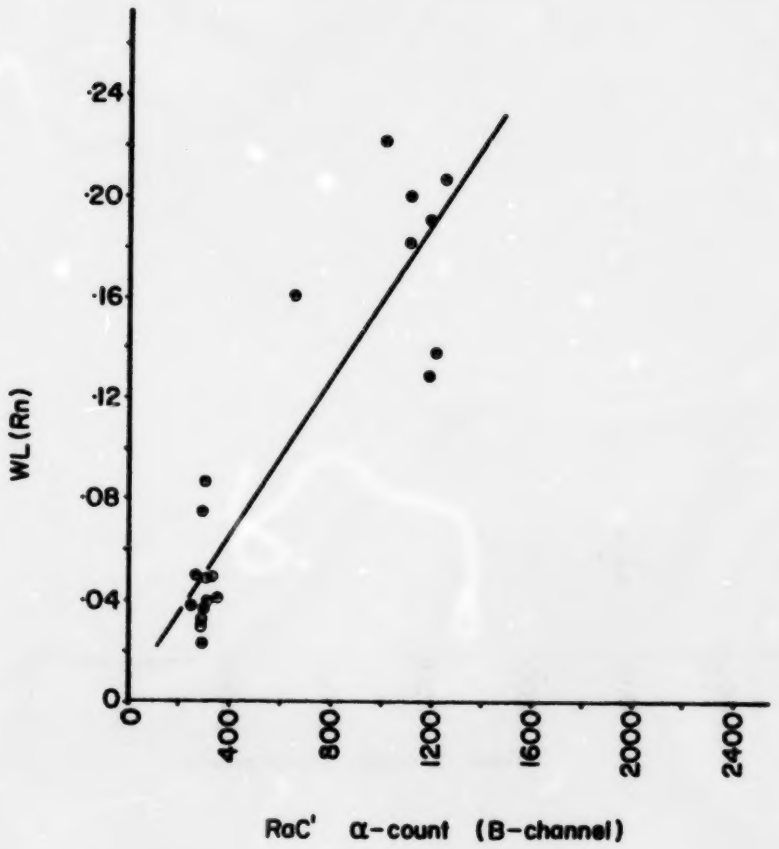

Fig. 11 - Dosimeter RaC' α-count versus Working Level.
Dosimeter reading taken 3.5 h after sampling.

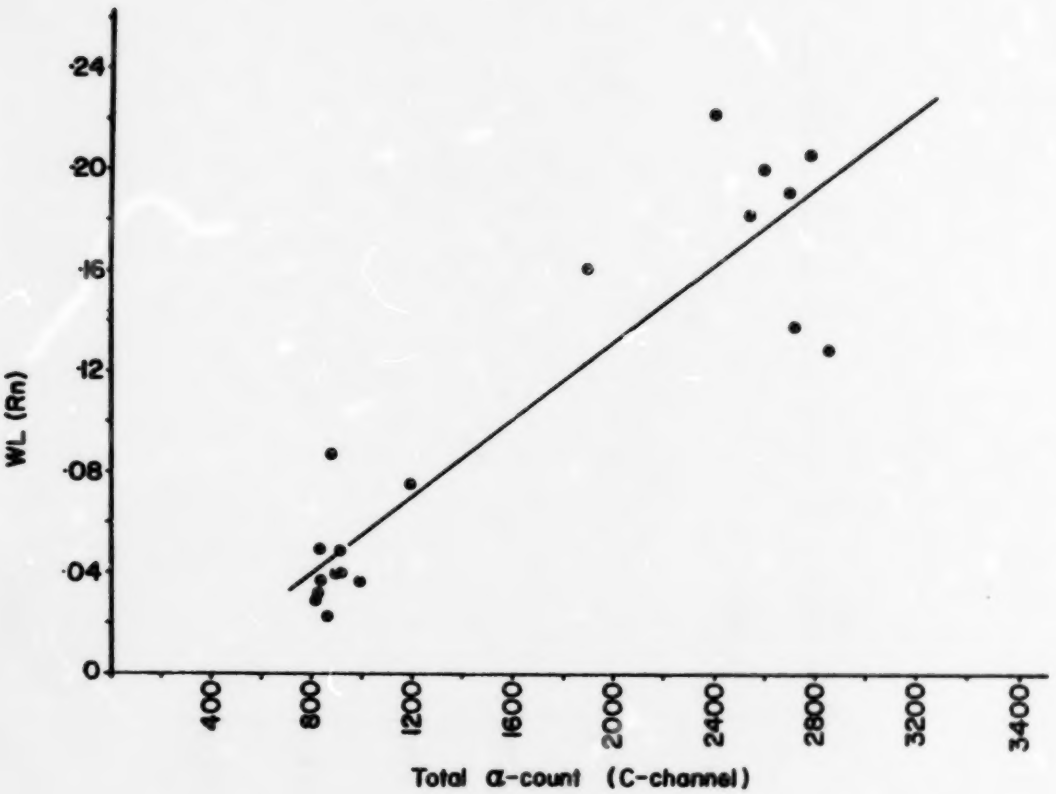

Fig. 12 - Dosimeter total α-count versus Working Level.
Dosimeter reading taken at the end of sampling.

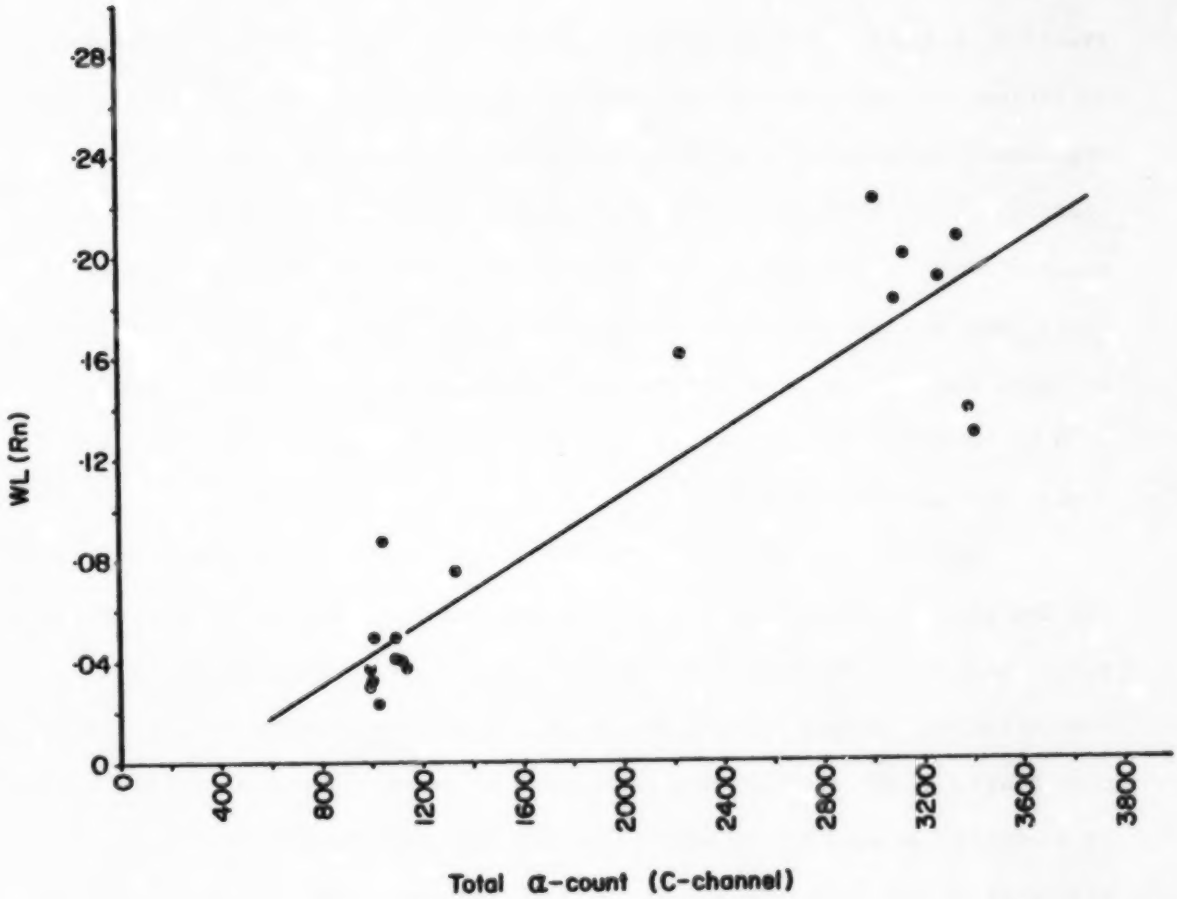

Fig. 13 - Dosimeter total α-count versus Working Level.
Dosimeter reading taken 3.5 h after sampling.

are summarized in Table 1.

The relative high correlation coefficient (up to >0.99) obtained in linear regression analyses of α-count versus [Rn] indicates that for all practical purposes, one may assume a linear relationship between these two variables. A lower correlation coefficient (∿0.91) was obtained in linear regression analyses of α-count versus WL(Rn). This lower correlation coefficient is reflected in the wider spread of the experimental data, an observation which is partly attributed to the different Working Level Ratio (WLR) used in each test, i.e., from about 0.3 to 1.35 $mWL/pCiL^{-1}$. It should be noted that the range of theoretical values of WLR is 0 to 0.01 $WL/pCiL^{-1}$, or 0 to 10 $mWL/pCiL^{-1}$, derived from the definitions WLR = WL(Rn)/[Rn] and 1 WL(= 10^3 mWL)≡ 10^2 $pCiL^{-1}$.

Quite noticeable is the low WLR in the course of experimentation. The low experimental values for WLR are the result of our particular environmental testing conditions, i.e., low relative humidity and low aerosol concentration. Hence, considerable radon daughter plate-out in the RTTF walls took place, an effect which was compounded by further plate-out on the blades of a mixing fan used to minimize radon and radon daughter concentration gradients in the RTTF. It should be indicated that large changes in WLR can easily be induced without changing [Rn] appreciably. This is so because radon daughters plate-out easily in varying degree according to aerosol concentration and other environmental factors, whereas radon gas is not affected by these variables. Most of the experimental data relating α-count and WL(Rn) were mainly obtained using two well differentiated radon gas concentrations in which the relative humidity and aerosol concentration in the RTTF varied sufficiently to change the WLR, and hence WL(Rn), appreciably.

Best correlation was found between [Rn] and RaC' α-count followed by

Table 1 - Linear regression analysis data relating radon daughter α-count to radon gas concentration, [Rn], and radon daughter Working Level, WL(Rn).

Radon Daughter	Time of Measurement	Slope m	Intercept b	Correlation Coefficient CF	Remarks
RaA	E.S.	0.1758	-57.46	0.971	[Rn]
RaA	3.5 h	0.1535	-53.86	0.975	[Rn]
RaC'	E.S.	0.3074	10.97	0.992	[Rn]
RaC'	3.5 h	0.2208	9.92	0.994	[Rn]
RnD	E.S.	0.1133	-35.1	0.985	[Rn]
RnD	3.5 h	0.0916	-29.82	0.989	[Rn]
RaA	E.S.	0.1172	-32.5	0.921	WL(Rn)
RaA	3.5 h	0.1017	-32.68	0.908	WL(Rn)
RaC'	E.S.	0.2145	4.84	0.896	WL(Rn)
RaC'	3.5 h	0.1533	5.07	0.902	WL(Rn)
RnD	E.S.	0.0764	-20.19	0.915	WL(Rn)
RnD	3.5 h	0.0622	-18.43	0.914	WL(Rn)

(Slope values for RaA, RaC', RnD: $\times 10^{-3}$; Intercept values for RaA, RaC', RnD: $\times 10^{-3}$)

The symbol RnD indicates total radon daughter α-count, i.e., RaA + RaC'. E.S. and 3.5 h stand, respectively, for α-count taken at the end of the sampling period and 3.5 hours after sampling. The remarks column indicates the variable the radon daughter α-count has been correlated with. The [Rn] and WL(Rn) can be obtained from linear regression data and radon daughter α-count as follows:

$$Y = mx + b$$

where x is α-count, and Y stands for [Rn] or WL(Rn).

[Rn] versus total α-count, i.e., RaA + RaC'. Correlation between [Rn] and RaA α-count was found to be somewhat lower than that for RaC' α-count. Correlation between WL(Rn) and α-count was, as indicated above, appreciably lower than that between [Rn] and α-count.

Although the WL(Rn) has been correlated to α-count it should be pointed out that the radon gas concentration is a more meaningful variable than WL(Rn) for calibrating a passive dosimeter using a radon diffusing membrane. This is so because radon daughters are removed by the membrane and hence the WL(Rn) loses its meaning as the WLR and radon daughter disequilibrium ratio can vary quite appreciably for the same radon gas concentration. However, correlation of [Rn] with α-count is based on the following assumptions.

1. [Rn] in the sensitive volume of the dosimeter is the same as that corresponding to the testing location, e.g., RTTF;
2. Diffusion of Rn-222 through the dosimeter's membrane from the testing location (i.e., region of higher [Rn] to the sensitive volume of the dosimeter (lower [Rn]) is instantaneous; and
3. No back-diffusion takes place, i.e., no diffusion of Rn-222 occurs from the dosimeter to outside the sensitive volume of the dosimeter. This is the situation when the dosimeter is removed from the testing location (high [Rn]) to the laboratory for data retrieval, i.e., region of low [Rn].

None of the assumptions indicated in items 1 to 3 are strictly true as the diffusion of Rn-222 through a membrane takes a finite time which depends on the characteristics of the membrane and the [Rn] gradient between outside and inside the dosimeter's sensitive volume. If the diffusion time of Rn-222 through the membrane is more than the sampling period, appreciable [Rn] gradients will be present. Large errors may also be introduced in studying the response of the dosimeter if the Rn-222 diffusion time in the membrane is an appreciable

fraction of the sampling period. The same observations apply when the dosimeter is removed from the sampling location to the laboratory for data analysis. In this case, appreciable uncertainty in the estimation of [Rn] arises when α-count is taken up to 3½ h after the end of the sampling period.

The data presented in Fig. 2 to 13 and Table 1 indicate that the instrument is quite sensitive and suitable for personal monitoring in underground uranium mines and other working areas. It is also suitable for environmental monitoring. The sensitivity of the instrument based on an 8 h sampling time can be calculated from data in Table 1. An approximate value for the sensitivity of the dosimeter is given as 1/m. Hence, the range of values according to the radon daughter α-count used is 3 to 11 counts/pCiL^{-1} and 5 to 16 counts/mWL.

The present passive dosimeter eliminates some of the drawbacks characteristic of active dosimeters, including high power consumption and pump flow rate variability. Software changes are being introduced to further automate and enhance the capabilities of the instrument for field use.

ACKNOWLEDGEMENT

The passive dosimeter has been developed by Alpha-NUCLEAR (Toronto) under CANMET (Energy, Mines and Resources Canada) contract according to the Elliot Lake Laboratory technical specifications.

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